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Activity report

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August 20, 2008

Disclaimer

This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

Activity report

This report is aimed to show my activities to support the LDRD.

Title: Investigation of the Double-C Behavior in the Pu-Ga Time-Temperature-Transformation Diagram

PI: Kerri Blobaum

Time: January – July, 2008

Prepared by Sung Woo Yu

Contents

This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. LDRD tracking #: 07-ERD-047.

1. Sample Holder Test

The final sample holder for Pu shown in Figure 1 is designed by Mark Wall. In order to make sure that the Pu sample is well separated from the air contamination not only at room temperature, but also at liquid nitrogen temperature, the following 5 tests are performed by Sung Woo Yu and Jason Jeffries.

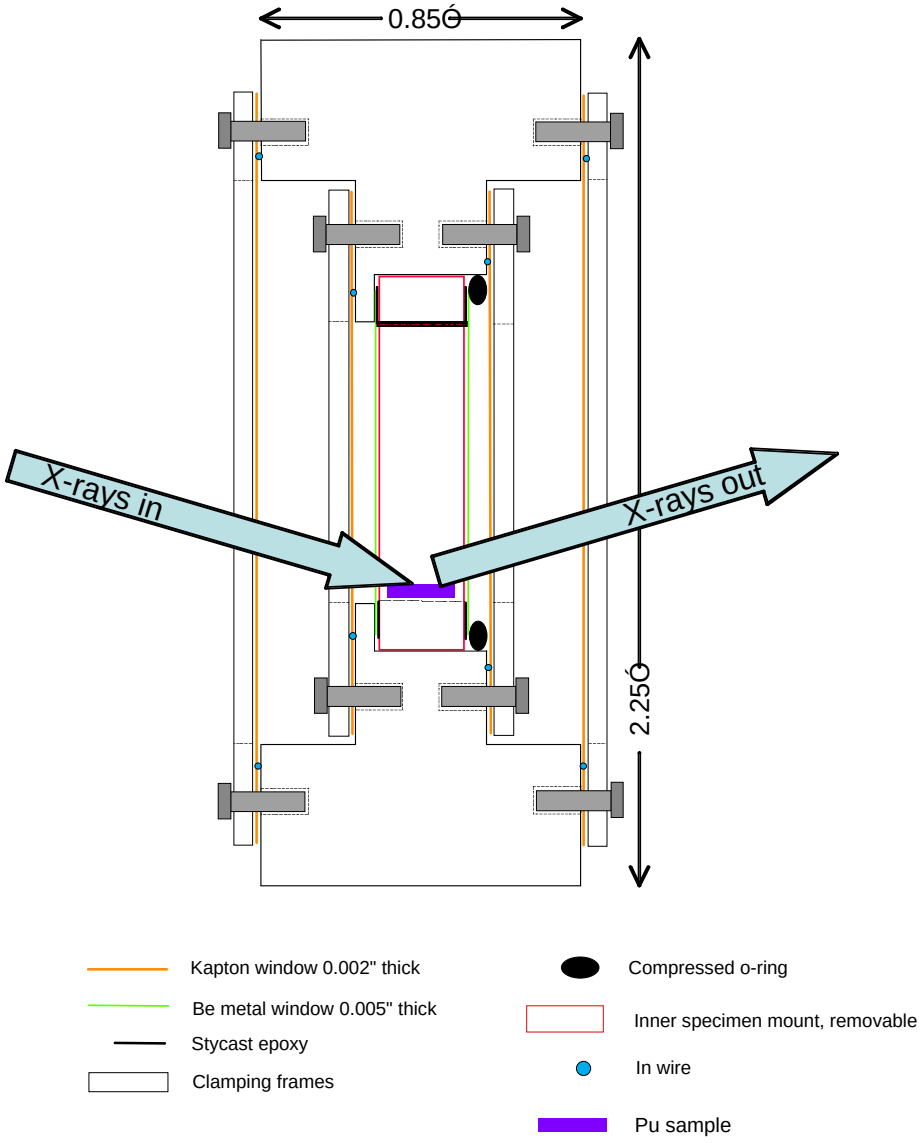


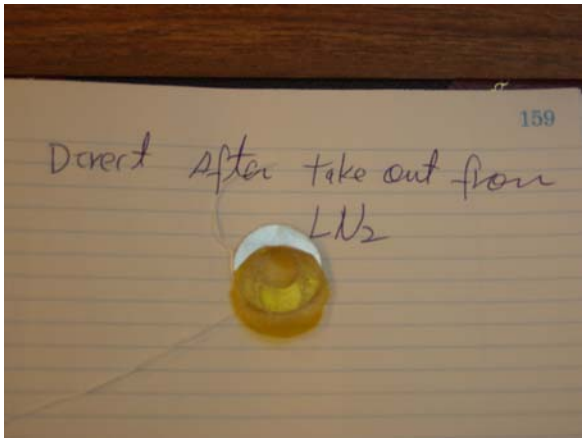
Figure 1: The Final Sample holder for Pu

Test 1: Stickiness Test with Kapton Tape

In order to test whether or not the Kapton tape can be used to seal the Pu sample not only at room temperature, but also at liquid nitrogen (LN2) temperature, the following tests have been performed.

1. Kapton tape is taped on the clean surface of a washer.
2. The washer is put into LN2 for 1 minute and the washer is taken out from LN2.
3. Tape is free from the washer. Kapton tape is stuck no more on the washer during the washer and Kapton tape were still cold).
4. After about 3 minute, the temperature of the washer and Kapton were almost RT, then, Stickiness comes back (no difference from step 1).
5. Steps 1-4 are repeated 5-6 times and same results are found.

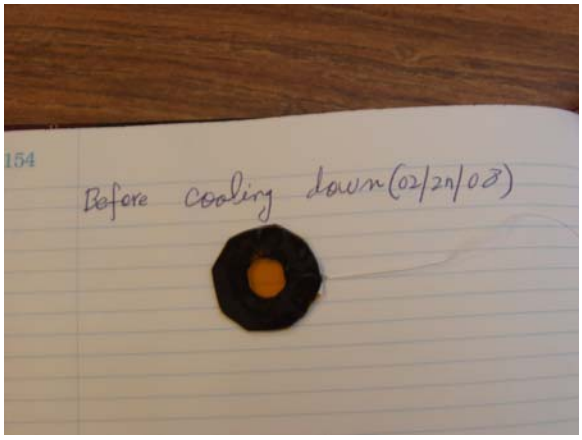
The conclusion is that the Kapton tape can not be used for the Pu sample holder.

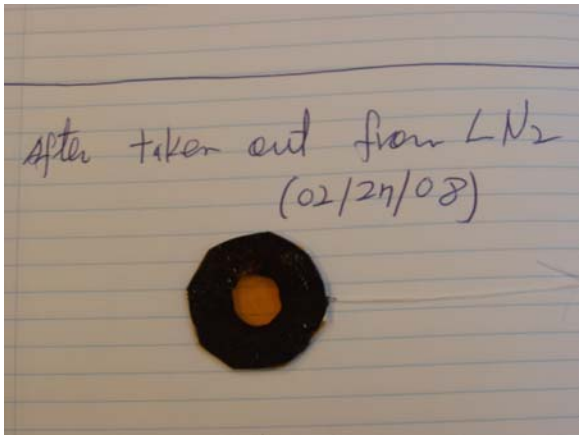


Test 2: Stickiness Test with Epoxy

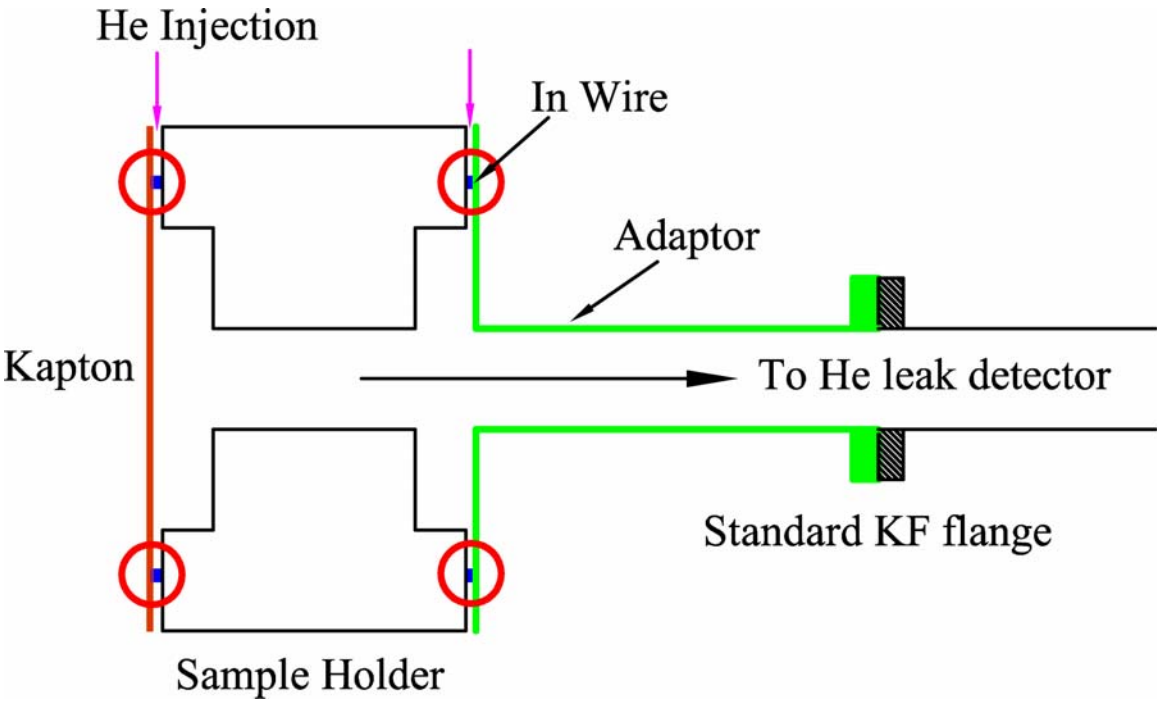
1. Kapton film is sticked on AL washer by STYCAST Epoxy (PART #: ES-2-20).
2. It is cured at RT for 45 hours.
3. Then, it is tested in LN2.
4. It is put into LN2 for 2 min and taken out and tested its stickiness. Stickiness doesn't change.
5. #4 is repeated for 5 times. Stickiness doesn't change.
6. It is put into LN2 for 20 min and taken out and tested its stickiness. Stickiness doesn't change.
7. # 6 is repeated for 4 times. Stickiness doesn't change.
8. It is put into LN2 for 2 hours and taken out and tested its stickiness. Stickiness doesn't change.

Conclusion: We found that STYCAST Epoxy works well to stick Al washer and Kapton film at LN2 temperature.





Test 3: Sealing with indium wire



Pre-condition

1. One side of the sample holder is covered with Kapton film (0.005 inch in thickness) with indium wire (0.02 inch in diameter).
2. Another side of the sample holder is connected to the He leak detector via the adapter (shown in green in the above sketch). The sample holder and the adaptor are sealed by the identical indium wire. The adaptor and the He leak detector are connected through standard KF flange.
3. After several hours pumping, the entire system was stabilized in pressure and the leak rate was $3x10^{-7}$ atmospheric-cc/sec (atm-cc/sec). The partial pressure in the test chamber of the leak detector was better than $1x10^{-3}$ torr.

Test 1

4. I injected very small amount of He gas to indium wire as shown in the above sketch as pink arrow, and I tried to find any leaks through the indium wire. Very slowly I checked the entire indium wires. After several time trials, I found NO Leak. It is very important to note that we have to use a very small amount of He gas in order to find a leak around the indium wire. For example, if we inject some amount of He gas, He atoms move everywhere and some He atoms go into the inside of the sample holder through the Kapton. (Because of nonzero permeability of He, He atoms pass through the Kapton). Very open, it causes wrong information that we had a leak through the indium wire.

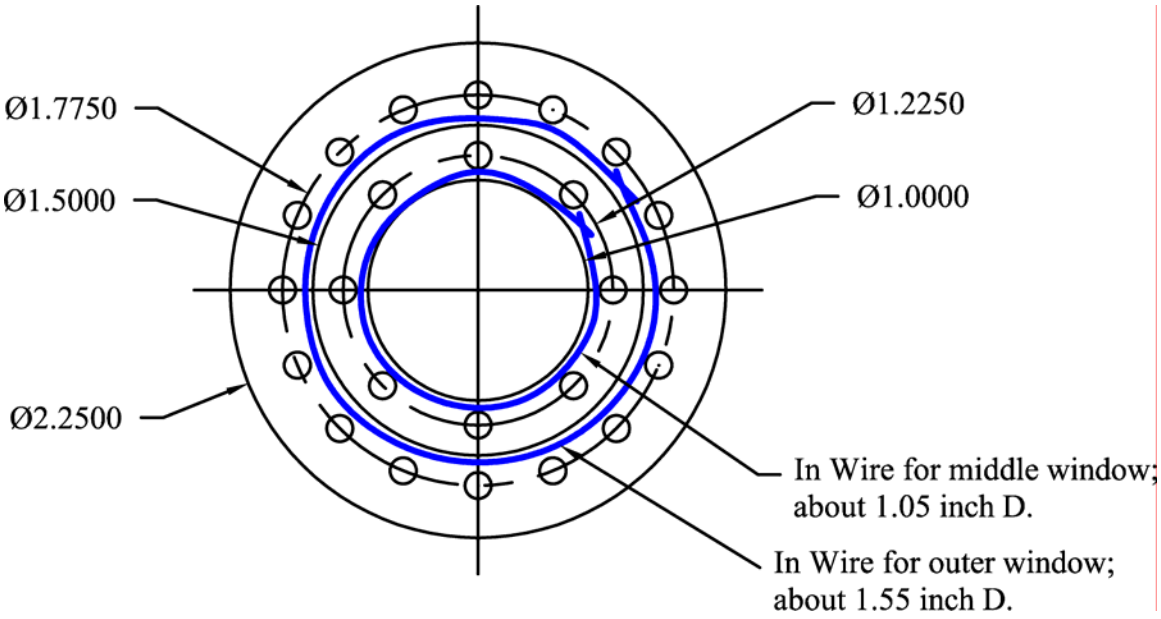
5. The entire sample holder is embedded in LN2 for 20 minute. During the period of the embedding, there was no change in the leak rate. After the sample holder was taken out from LN2, step 4 is repeated. I found NO leak.
6. Steps 4 and 5 are repeated for 3 times.
7. The entire sample holder is embedded in LN2 for two hours. After the sample holder was taken out from LN2, step 4 is repeated. I found NO leak.
8. The sample holder is disconnected at the standard KF flange and the KF flange is covered with a blind KF flange. It doesn't change the leak rate in the leak detector.
9. Steps 1-8 did also for the middle window (0.002 inch in thickness).

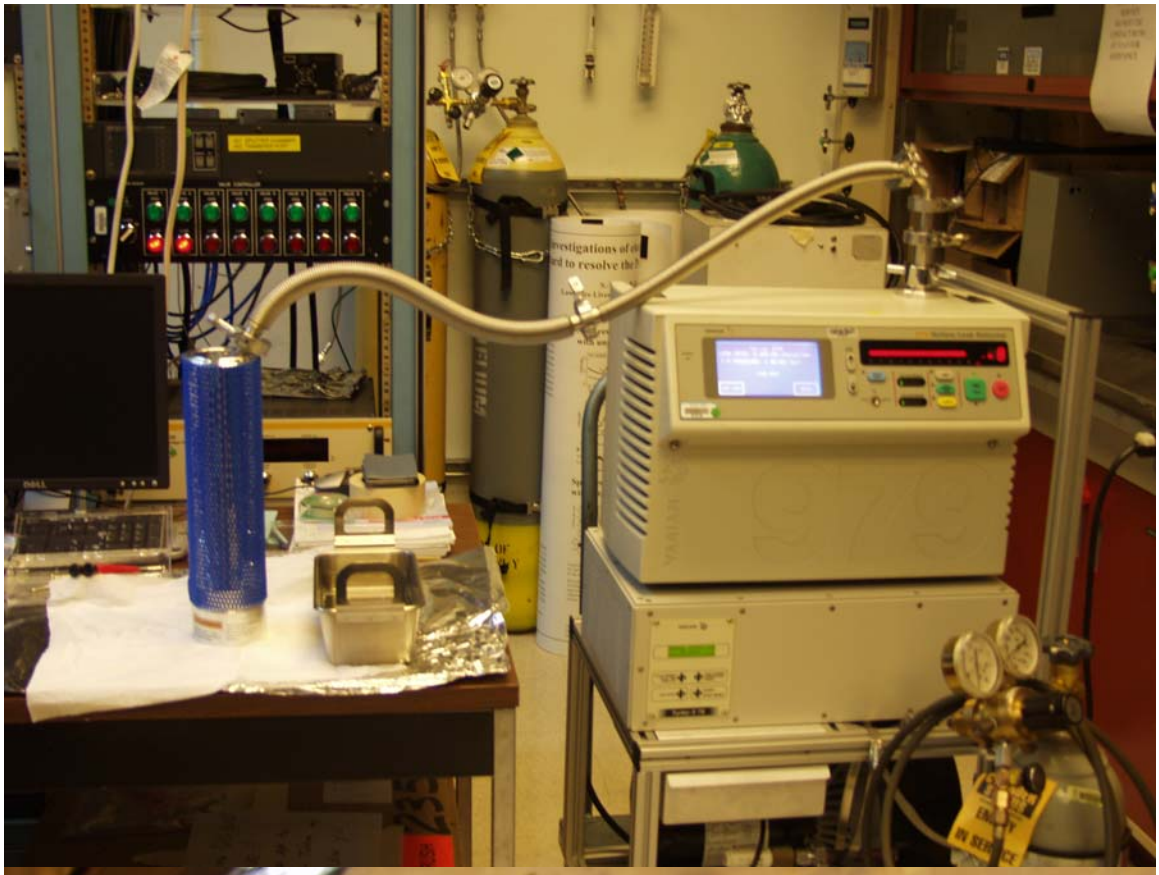
Test 2: the leak rate is $1x10^{-7}$ atom-cc/sec

10. The entire sample holder is embedded in LN2 for about 3 minutes, which is the time the sample holder needs to be cooled down to LN2 temperature. After that, the sample holder is embedded into RT water for about 3 minutes, which is the time the sample holder needs to come back to RT.
11. Step 10 is repeated for 30 times. After every 10 times of step 10, I tried to find a leak with step 4. I found No leak.
12. The sample holder is disconnected at the standard KF flange and the KF flange is covered with a blind KF flange. It doesn't change the leak rate in the leak detector.
13. Steps 10 and 12 did also for the middle window (0.002 inch in thickness).

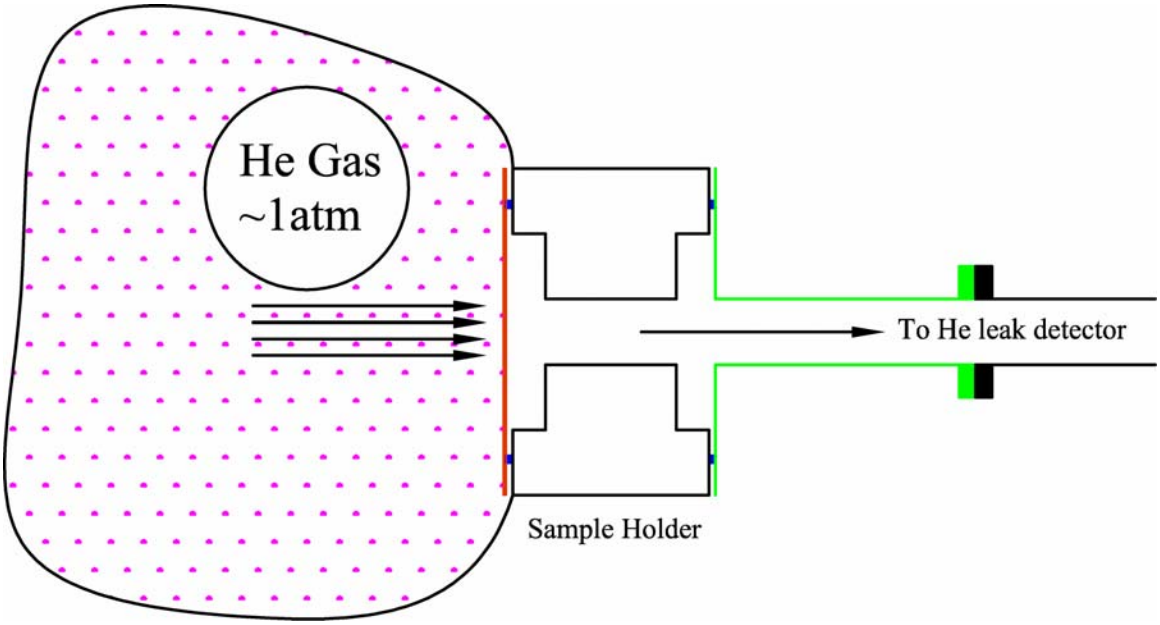
Conclusion

Indium wire (0.02 inch in diameter) works well to seal the sample holder and the Kapton for vacuum at RT and at LN2 temperature.





Test 4: Additional test to check the permeability of the system as a whole



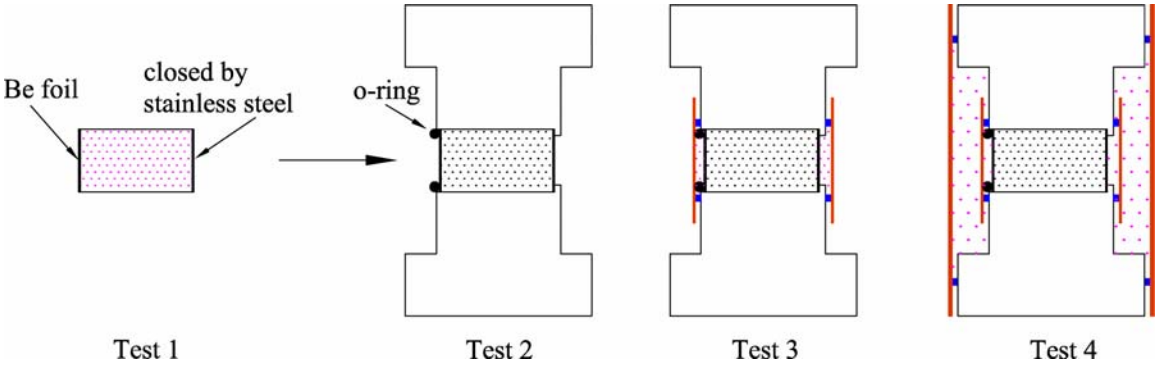
Pre-condition

14. We taped a bag around the sample holder when it is attached to the leak detector as shown in the above sketch. The bag is filled with ~1 atm of helium gas.

Test 1

15. We measured the leak rates for the outer window for 45 minute and for the middle window for 45 minute.
16. The leak rate for the outer window is 1.2x10-5 atm-cc/sec
17. the leak rate for the middle window is 1.4x10-5 atm-cc/sec

Test 5: Additional test to check the mechanical stability of the Be window under vacuum and the permeability of the system as a whole



Test 1

1. The inner sample holder (it will hold Pu sample directly) is sealed by Be foil (0.005 inch in thickness) on the one side with silicone and is closed by stainless steel on the other side under He environment. This means that the inside of the inner sample holder contains about 1 atm He gas.
2. Then the inner sample holder is placed in the desiccator which is connected to the leak detector. The leak detector pumped on the desiccator and the leak rate is measured. The He leak rate through the Be window was about $1x10^{-6}$ atm-cc/sec.

Test 2

3. This time, the inner sample holder is sealed according to step 1, but under air. This means that the inside of the inner sample holder contains about 1 atm air.
4. Then the inner sample holder is inserted into the sample holder and then fixed with the o-ring and the screws.
5. Then the sample holder is placed in the desiccator which is connected to the leak detector. The leak detector pumped on the desiccator and the leak rate is measured. The leak rate was about $3x10^{-9}$ atm-cc/sec.
6. Then the sample holder is removed from the desiccator and the leak rate is measured without the sample holder. The leak rate was identical to step 5.
7. The main reason for tests 1 and 2 is to see any mechanical failure under vacuum on the Be window sealed with silicon epoxy. Therefore, we visually checked the Be window carefully after the step 5. We didn't see any deformation.

Test 3

8. The middle windows are sealed with Kapton film using indium wire under He environment.
9. Then the sample holder is placed in the desiccator which is connected to the leak detector. The leak detector pumped on the desiccator and the leak rate is measured. The leak rate was about $1.5x10^{-5}$ atm-cc/sec. This leak rate is

Test 4

10. The outer windows are sealed with Kapton film using indium wire under He environment.
11. Then the sample holder is placed in the desiccator which is connected to the leak detector. The leak detector pumped on the desiccator and the leak rate is measured. The leak rate was about 1.6×10^{-5} atm-cc/sec.

Based on the above 5 tests, Kerri Blobaum made the final document for IWS#14387. This IWS is approved for the experiments at the APS.

2. Calculation of diffraction patterns

I have calculated the x-ray diffraction patters for all the possible phases of Pu with photon energies of (8.2keV, 17.7keV, 20keV, 40keV, and 60keV) to see:

1. How the 2 theta (2θ) changes as a function of photon energy. As shown in Figure 2, the cryostat has a maximum theta of 13°, this means 2θ=26°. When we take a look at the diffraction pattern with 20keV, which is the photon energy we will use at the APS, the 2 theta (2θ) = 14° for the primary diffracted patterns. This means that the experimental geometry with cryostat is OK.
2. To distinguish between α-Pu, δ-Pu, γ-Pu in diffraction patterns.

I also have been trying to calculate the diffraction patterns for the Pu-Ga system. Initially Kerri Blobaum informed me to use GSAS to calculate the diffraction patterns for Pu-Ga system, but it is turned that GSAS was not an appropriate program for this. According to the Hyunchae Cynn (High Pressure Physics Group) and Nobumichi Tamura (Lawrence Berkeley National Lab), I have been using POWDERCELL. Progress is being made, but a problem is that the POWDERCELL doesn't allow me to change the lattice parameters and the number of atoms for unit cell. More time has to be spent to resolve the problem.

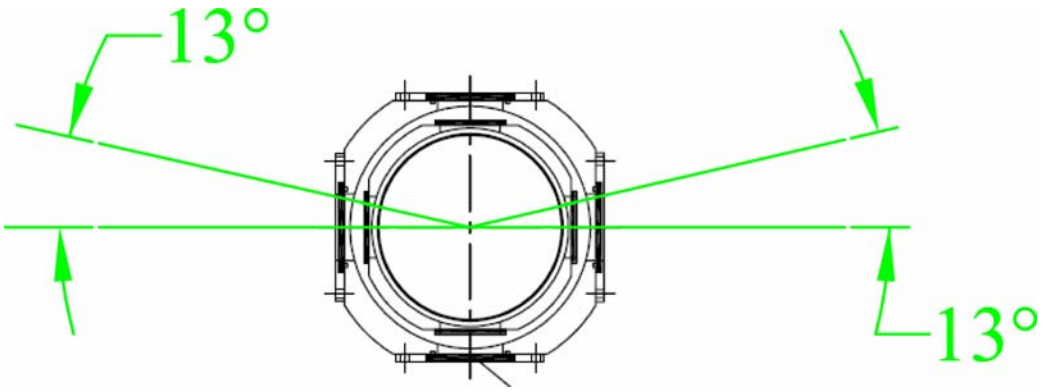
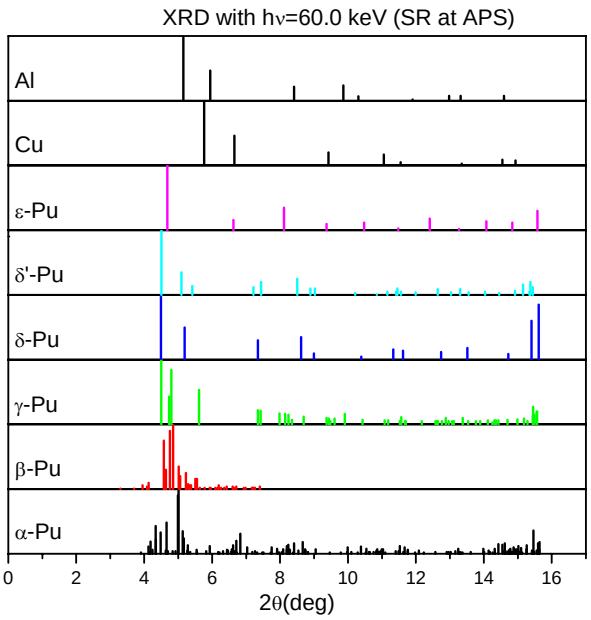
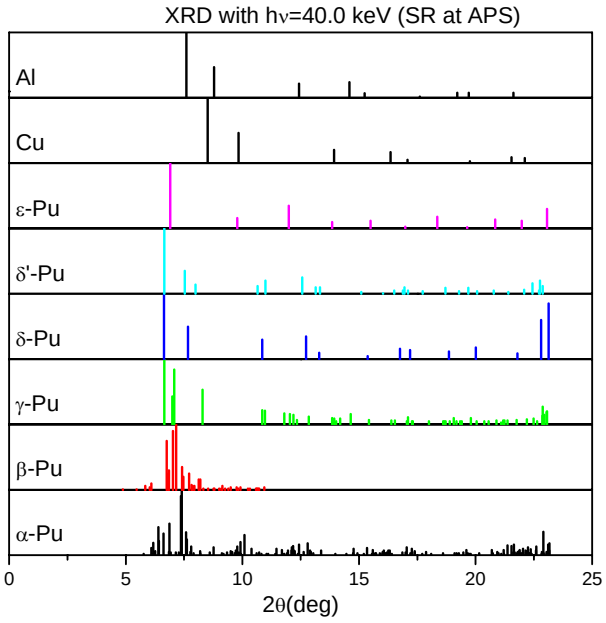


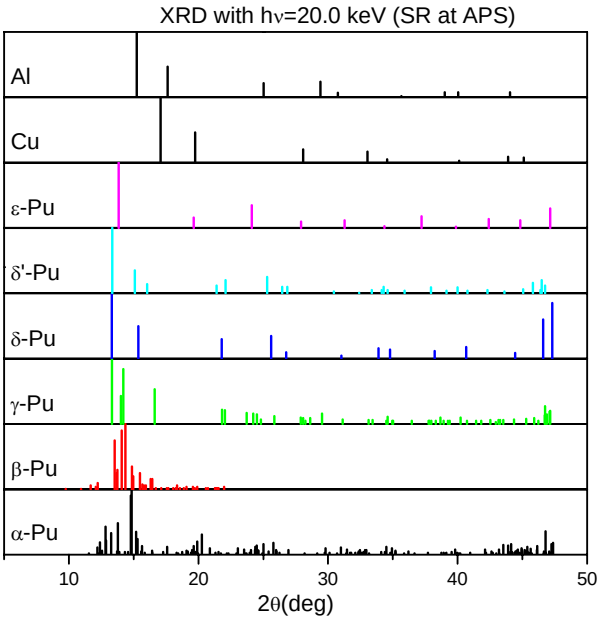
Figure 2: Experimental geometry with the cryostat.



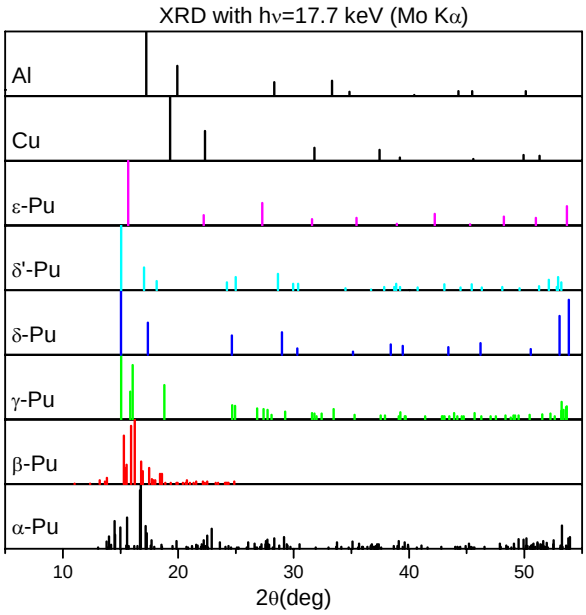
XRD with hν=60.0 keV: These XRD is calculated by SWY.



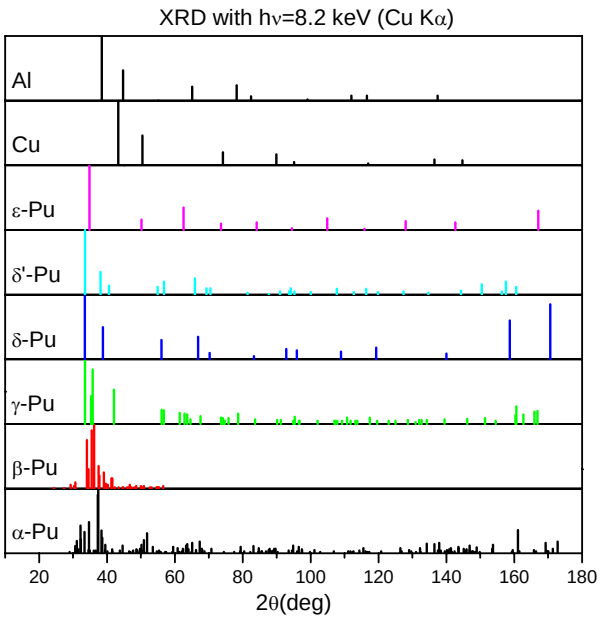
XRD with hν=40.0 keV: These XRD is calculated by SWY.



XRD with hν=20.0 keV: These XRD is calculated by SWY.



XRD with hν=17.7 keV: These XRD is calculated by SWY using the XRD with hν=8.2 keV and the diffraction equation.



XRD with hν=8.2 keV: Cheng Saw provided the data from his database and SWY plotted.

3. Literature search and preparing publications

I have spent considerable time to find papers for plutonium. I have read approximately 50 papers for Pu and Pu alloy systems to extend my knowledge about Pu. Based on those papers, I have prepared a very rough draft for a publication, even though we haven't got the experimental diffraction data from Pu-Ga system yet.

I also learned the computer software for the calculation of x-ray diffraction pattern: GASA and POWDERCELL.

Paper lists:

1. Hecker et al, Progress in Materials Science **49**, 429 (2004).
2. Blobaum et al, Acta Materials **54**, 4001 (2006).
3. Blobaum et al, Metallurgical and Materials Transaction A, **37A**, 567 (2006).
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8. Abramenko et al, J. Alloys and Compounds **444-445**, 207 (2007).
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11. Massalski et al, J. Alloys and Compounds **444-445**, 98 (2007).
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13. Ellinger et al, Journal of Nuclear Materials **12**, 226 (1964).
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21. Hirth et al, Acta Materials **54**, 1917 (2006).
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35. Wong et al, Science **301**, 1078 (2003).
36. Wong et al, PRB **72**, 064115 (2005).
37. Soderlind et al, PRL **92**, 185702.
38. Lookman, PRL **100**, 145504 (2008).
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40. Savrasov et al, Nature **410**, 793 (2001).
41. Tobin et al, PRB **72**, 085109 (2005).
42. Moore et al, Philosophical Magazine **84**, 1039 (2004).
43. Baptist et al, J. Phys. F : Met. Phys. **12**, 2103 (1982).
44. Shim et al, Nature **446**, 513 (2007).
45. Pourovskii et al, EPL **74**, 479 (2006).
46. Shorikov et al, PRB **72**, 024458 (2005).
47. Tobin, Journal of Alloys and Compounds **444-445**, 154 (2007).
48. Tobin et al , J. Phys.: Condens. Matter **20**, 125204 (2008).
49. Julien et al, PRB **77**, 195113 (2008).
50. Batista et al, submitted to PRL (2008), LA-UR-08-0786
51. Shick et al, EPL **69**, 588 (2005).
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4. Tasks Required for APS Experiments

The following table shows the tasks required for APS experiments. Based on this, I did my responsibilities diligently, communicating with other collaborators (Jason Jeffries, Technicians, Kerri Blobaum, Will Evans and other people in Physics).

Task	Responsible person	Due date
Initial tests of Stycast epoxy (or other adhesive) through cold temperature cycles	Mark Wall, Sung Woo Yu	May 9, 2008
Test beryllium foil with stycast epoxy through cold temperature cycles (including written documentation)	Mark Wall (documentation)	May 23, 2008
Order sample holder supplies (Stycast, indium wire, o-rings, beryllium foil, etc.) Cut Kapton windows	Mark Wall, Sung Woo	May 23, 2008
Calculate pressure on windows when the sample holder is in the cryostat vacuum	Kerri, Sung Woo	May 16, 2008
Determine pressure rating for Kapton (DuPont should have specs)	Sung Woo	May 16, 2008
Test sample holder in vacuum (including written documentation)	Sung Woo	May 23, 2008
Test sample holder cold in vacuum through thermal cycles (including written documentation)	Sung Woo	May 30, 2008
Obtain limits on amount of material from APS (in process)	Will Evans	May 23, 2008
Fabricate mechanism for mounting samples	Mark Wall	May 30, 2008
Test mechanism for mounting samples through cold cycles (including written documentation)	Jason, Mark, Sung Woo	June 6, 2008
Install thermocouple in sample holder	Jason, Sung Woo	June 27, 2008

Complete wiring in cryostat	Jason, Sung Woo	June 27, 2008
Fabricate bracket for holding sample holder in cryostat (including modifications to sample holder)	Mark Wall	June 27, 2008
Full tests of cryostat with temperature controller (setting PID, etc.)	Sung Woo, Jason	July 3, 2008
Measure cooling/heating rate in cryostat	Jason	July 8, 2008
Full test of experimental conditions in cryostat (no Pu)	Jason, Sung Woo	July 10, 2008
IWS written, ready for authorization	Kerri Blobaum	June 13, 2008
Obtain safety authorization from APS	Will Evans	June 13, 2008
Ship cryostat to APS	Will Evans	July 14, 2008
Load sample, ship to APS	Mark Wall	July 25, 2008
Collect data at APS		August 2-6, 2008

Scientific Tasks

Task	Responsible person	Due date
Simulate XRD patterns for α', δ, and γ' for a Pu-2.0 at% Ga alloy using GSAS software		May 30, 2008
Simulate XRD patterns for α', δ, and γ' at cryogenic temperatures		June 13, 2008
Determine thermal path for experiments		July 3, 2008
Collect data at APS		August 2-6, 2008

5. Communications

Regarding this project, I communicated with collaborators via email, phone, direct conversations and meetings:

1. Email: I exchanged about 100 emails (70 with Kerri Blobaum, 13 with Jason Jeffries, 10 with Mark Walls, 4 with Will Evans, 2 with Ken Visbeck……)
2. Phone and direct conversations: Kerri Blobaum, Jason Jeffries, Mark Wall, Cheng Saw, Jim Tobin, Richard Gross, Ray Swan, Larry Walkley, Hyunchae Cynn (High Pressure Physics Group), Jae-Hyun Park Klepeis (High Pressure Physics Group), Nobumichi Tamura (Lawrence Berkeley National Lab).
3. Several Meetings